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Near-Infrared Lasing at 1 µm from a Dilute Nitride-Based Multishell Nanowire

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ABSTRACT: A coherent photon source emitting at near-infrared (NIR) wavelengths is at the heart of a wide variety of applications ranging from telecommunications, optical gas sensing to biological imaging, and metrology. NIR-emitting semiconductor nanowires (NWs), acting both as a miniaturized optical resonator and as a photonic gain medium, are among the best-suited nanomaterials to achieve such goals. In this study, we demonstrate the NIR lasing at 1 µm from GaAs/GaNAs/GaAs core/shell/cap dilute nitride nanowires with only 2.5 % of nitrogen. The achieved lasing is characterized by an “S”-shape pump-power dependence and narrowing of the emission line-width. Through examining the lasing performance from a set of different single NWs, a threshold gain, $g_{th}$, of 4100 – 4800 cm$^{-1}$, was derived, with a spontaneous emission coupling factor, $\beta$, up to 0.8, which demonstrate the great potential of such nanophotonic material. The lasing mode was found to arise from the fundamental HE$_{11a}$ mode.
of the Fabry-Perot cavity from a single NW, exhibiting optical polarization along the NW axis. Based on temperature dependence of the lasing emission, a high characteristic temperature, $T_0$, of 160($\pm$10) K is estimated. Our results, therefore, demonstrate a promising alternative route to achieve room-temperature NIR NW lasers thanks to the excellent alloy tunability and superior optical performance of such dilute nitride materials.
Nanoscale photon sources with coherent light emission in the near-infrared (NIR) spectral range are key elements for a host of technologically important applications, ranging from optical data communication to gas sensing and biological metrology. Semiconductor nanowires (NWs), with their naturally formed Fabry-Perot (F-P) cavity and an optically active medium enabling light amplification, are widely considered as promising candidates for realizing the miniaturized NIR laser with subwavelength footprint, ideally for on-chip integration with low power consumption and superior optical performance. So far, the most common semiconductor materials employed for the NIR nanowire lasers are based on GaAs [1 – 11], (In, Ga)As [12 – 20], and (In, As)P alloys [21 – 26]. Pushing the lasing wavelength further into IR region usually requires a higher alloying composition of the narrow-gap semiconductor component. For example, an In content of at least 15 - 20 % is required in InGaAs quantum dot (QD), quantum well (QW) and uniform NWs to give a lasing emission beyond 950 nm [12 – 20]. The InAsP QD-embedded NW lasers, which emit at the telecom window of 1.3 µm, contains an As composition up to 60 % [26]. Recently, lasing radiation at 990 nm was demonstrated in GaAsSb superlattice NWs [27] with 8 % of Sb. Lattice mismatch between the optical gain medium and surrounding barrier/passivation materials gives rise to strain across the NW structure, which consequently modifies and complicates the electronic states of the emitter and the resulting optical properties. Therefore, a semiconductor alloy with both exceptionally large bandgap tunability in the NIR spectral range and a minimized lattice misfit with its parental counterpart is highly desired to construct opto-electronic devices.
Dilute nitride GaNAs has in recent years emerged as a promising candidate for the NIR nanophotonic applications. Thanks to the giant bandgap bowing effect induced by the strong anticrossing interaction between the conduction band (CB) of GaAs and the localized energy level of nitrogen (N) [28 – 31], the CB minimum of the forming alloy dramatically shifts down in energy. This results in a significantly reduced bandgap of GaNAs, e.g. the bandgap is reduced by 260 meV in the alloy with only 2 % of N. The GaNAs band gap energy exhibits better thermal stability than that in the parental GaAs [32 – 34], which results in a thermally stable spectrum of the emitted light as required for optoelectronic devices. Furthermore, it was found that the presence of N can partially inhibit the formation of nonradiative surface states in GaNAs and GaNP nanostructures [35-38], leading to a prolonged lifetime of photo-excited carriers. On the other hand, the formation of point defects, such as Ga interstitials, facilitated by N incorporation allows to achieve efficient spin filtering [39] and spin amplification [40] in these materials even at room temperature. Based on these excellent material properties, a variety of novel applications have been demonstrated in GaNAs-based nanopillars. For example, we have recently demonstrated that GaNAs nanopillar structures provide an efficient spin-photon interface featuring both a high light-coupling efficiency and a record-high room-temperature spin polarization degree up to 60 % [40], promising for future spin-optoelectronics-based quantum information technology. We have also achieved lasing from a single GaNAs NW, which emits at 875 nm with an N content [N] of 0.5 % [41]. Extending their performance towards longer wavelength is essential for the realization of NIR coherent photon sources. Here,
we report on the NIR lasing at 1 µm from a GaAs/GaNAs/GaAs core/shell/cap NW with only 2.5 % of N. By controlling the thickness of the GaNAs shell, thus the cross-sectional area of the optical gain medium, the origin of the lasing mode can be tailored. We observe lasing via the fundamental HE$_{11a}$-mode for the 50 nm-thick GaNAs shell, whereas it switches to the HE$_{21b}$ mode when the shell thickness increases to 100 nm [41]. The lasing emission can be sustained up to 100 K, which is primarily limited by the pump fluence of our laser excitation at the higher temperatures. A characteristic temperature, $T_0$, of 160(±10) K is derived, which is comparable with the previously reported values for room-temperature NW lasers [7, 12] and higher than $T_0 = 90$ K for the bare GaAs/GaNAs core/shell wires without surface passivation [41]. Our work, therefore, presents an important advance towards exploiting dilute nitride GaNAs as a gain material for the NIR-emitting NW lasers beyond 1 µm, with outstanding flexibility in laser mode profiling.

The investigated GaAs/GaNAs/GaAs core/shell/cap NWs have a GaAs core of 200 nm in diameter, followed by a 50-nm thick GaNAs shell, and finally covered by a 50-nm thick GaAs cap layer. The cross-section of the NWs is illustrated in the lower part of Figure 1b. For photoluminescence (PL) spectroscopic studies of single NWs, we transferred the wires onto a gold substrate for better thermal conductivity under optical pumping (see Methods for details about the NW growth and the PL setup). It has been found in our previous study that the gold substrate will not affect the PL dynamics in NWs due to coupling with surface plasmons (SPs) [41], as the wires have a larger size than the mode volume of the localized SPs. Therefore, the
observed PL signals reflect the pure photonic character of the NWs. Shown in Figure 1a is a representative scanning electron microscopy image of the GaNAs NW, which we label as NW1. The NW has a length, $L$, of 6.3 µm, featured with clear and smooth sidewalls enabling good confinement of optical modes in the Fabry-Perot cavity. Only slight tapering is observed along the NW. Therefore, we treat it as uniform with the diameter of 400 nm in the theoretical simulations to be presented below.

Alloying homogeneity is pivotal for the gain quality of an active medium, as it allows a uniform density distribution of optically/electrically pumped carriers for optimal light amplification. Given the giant bandgap bowing effect upon N incorporation, an evaluation of the uniformity in N composition along the wire is necessary. For this purpose, we performed room-temperature spatially resolved PL measurements on single NWs and the corresponding results for NW1 are shown in Figure 1b. PL spectra were recorded by scanning the excitation along the wire axis from the bottom to top, corresponding to the positions ‘1’ to 11’ shown in Figure 1a. The PL spectra are dominated by a broad PL band centered at around 1090 (1110) nm at the bottom (top) end of the NW, which stems from the GaNAs shell with the bandgap energy, $E_g$, of 1.138 (1.117) eV. At the higher energies the PL emission from the GaAs regions is also seen and peaks at around 1.42 eV, which corresponds to the GaAs bandgap. Its intensity is much weaker, indicating efficient carrier injection into the GaNAs shell. By invoking the band anti-crossing model to fit the $E_g$ of GaNAs [28, 29], the nitrogen composition is estimated
to be 2.5 (2.7) % at the NW bottom (top). The observed homogeneity of the alloy composition along the NW axis underscores the high quality of the material.

Temperature-dependent PL measurements were then carried out to evaluate its effect on the emission wavelength, and the corresponding results are displayed in Figure 1c and d. As temperature decreases, the PL emission blueshifts to the higher energy due to an increase in $E_g$. A weaker PL band appearing at 1.28 eV on the high-energy shoulder of the main emission peak is most likely due to unintentional incorporation of N into the GaAs core during the growth process, which has a lower N composition than the GaNAs shell and thus a larger bandgap energy. Such effect has been confirmed in our previous work. The PL emission from the GaAs core typically exhibits fast thermal quenching [42], as is also observed here for the 1.28 eV PL band. The thermally induced bandgap reduction is better visualized by normalized PL mapping shown in Figure 1d, where the PL maximum position as a function of temperature is marked by the dashed arrow. The bandgap changes by ~50 meV with varying temperature between 4 and 300 K. This value is appreciably smaller than ~90 meV typical for GaAs, hence reflecting better thermal stability of GaNAs as an effective gain material.

By taking a close look at the low-T PL spectra shown in Figure 1c, one notices clear periodic undulations that are the most visible on the low-energy side of the spectra. These undulations evidence coupling of the GaNAs emission into the optical cavity formed by the NW structure. To investigate the lasing capacity of the NW, we have studied its emission properties under pulsed laser excitation. The corresponding results obtained at 5K are depicted
in Figure 2a. With increasing excitation power fluence, $P_{\text{exc}}$, the high-energy PL edge of the NW1 exhibits a blue shift, owing to phase space filling of the continuum band by the photoexcited carriers/excitons. Upon $P_{\text{exc}}$ exceeding a threshold, $P_{\text{th}}$, of 15 µJ cm$^{-2}$ per pulse, several sharp PL lines, which are superimposed on the broad spontaneous emission (SE) background, appear. These sharp PL lines grow superlinearly in their intensity with increasing $P_{\text{exc}}$, marking the onset of the amplified spontaneous emission (ASE), and finally dominate over the whole PL emission with a nearly 20 dB ratio to the SE background - a hallmark of lasing radiation. A full-width-at-half-maximum (FWHM) of 1.6 nm is extracted for the lasing line, yielding a cavity Q-factor of ~625. Lasing radiation is also demonstrated from wires with different lengths, $L$, as shown in Figure 2b and 2c for NW2 and NW3 with $L=4.4$ µm and $L=3.4$ µm, respectively (see also Supporting Information I). Based on the results of Figure 2a-c, mapping of the normalized PL intensity as a function of pump fluence is provided in Figure 2d-f, for clear visualization. We summarize the excitation power dependences of spectrally integrated PL intensities for the lasing lines labeled as ‘i’-‘iii’ in a light input-light output (L-L) plot, - see the solid symbols in Figure 2g. The characteristic ‘S’-like power dependence was observed for all the sharp lines, showing the evolution of radiation processes from SE, to ASE and to lasing, which is accompanied by a simultaneous narrowing of linewidth (the open symbols in Figure 2g). These changes of the PL origin are also reflected by corresponding changes in polarization of the emission. By taking NW2 as an example, the lasing line shows the pronounced polarization along the NW axis as depicted by the solid dots in the inset of Figure 2b. This is in
sharp contrast to the unpolarized PL background in the SE regime represented by the open circles. We note that the optical antenna effect, which causes polarization of the NW emission along the wire axis in thin NWs, is vanishingly small in our case, as the NW diameter is well beyond the sub-hundred nm regime where the antenna-induced optical anisotropy prevails [43, 44]. We employed a rate equation analysis to quantitatively interpret the power dependence of the lasing process (see Supporting Information II), based on which a threshold gain, $g_{\text{th}}$, of 4200 cm$^{-1}$, 4800 cm$^{-1}$, and 4100 cm$^{-1}$ was derived for NW1-3. The best fits to the experimental data are shown by the solid lines in Figure 3g. It is noteworthy that whereas NW1 and 2 exhibit an apparent ASE kink correlated with the deduced SE coupling factor, $\beta$, of 0.065 and 0.02, the threshold for NW3 is less pronounced, rendering $\beta \sim 0.8$. We note the high $\beta$ value for NW3 can be ascribed to several factors. Firstly, NW3 exhibits stronger PL emission as shown in Figure 2c, which implies a higher radiative efficiency due to e.g. better crystalline quality and is corroborated by temperature-dependent PL measurement (see Supporting Information III). Secondly, the higher $\beta$ value for NW3 may be partly related to an improved quality of the cavity. As is reflected by the SEM images of NW1-3 given in Figure 1 and Figure S1 in Supporting Information I, the end facets of NW1 and NW2 are less well-defined as compared with NW3, i.e. the bottom end of NW1 shows split side branches and NW2 exhibits larger tapering in diameter where the top part is not sharply cone-like. Therefore, NW3 possesses the best cavity among all, which is also consistent with its lowest lasing threshold. In addition, we considered the effect of a total number of longitudinal modes on $\beta$. In principle, fewer longitudinal modes
within the spectral gain bandwidth facilitates a higher $\beta$ [45, 46], as the spontaneous emission can more efficiently be coupled into the specific lasing resonance. To corroborate this, we measured lasing from several NWs with different mode spacings (see Supporting Information IV). Indeed, we found a trend of increasing $\beta$ with increasing mode spacing (or fewer modes within the gain spectra). It should be noted the lasing was also achieved from NWs on SiO$_2$ substrate, see Supporting Information V for details. The efficient coupling of incoherent radiative recombination into the lasing mode manifests the immense potential of the utilized GaNAs-based NW structure for thresholdless NW lasers.

Having demonstrated the lasing from the GaAs/GaNAs/GaAs core/shell/cap NWs, it is important to discern the modal origin of the stimulated radiation. We note that for NW1-3, there are sets of weak sharp resonances equally spaced on both sides of the main lasing peak. Such phenomena are typically expected for a Fabry-Perot cavity, which arises from different orders of a longitudinal mode. The mode spacing (marked by the paired arrows in Figure 2a-c) is expected to inversely relate with the wire length, which is indeed the case. We conducted statistical measurements over dozens of wires with different $L$ and plot the corresponding results as dots in Figure 3a, where the stars represent results from NW1-3. As expected [9, 13, 41, 47], the mode spacing exhibits a linear dependence on the inverse $L$ as:

$$\Delta \lambda = \left( \frac{\lambda^2}{2L} \right) \left( n_{\text{eff}} - \lambda \frac{dn_{\text{eff}}}{d\lambda} \right)^{-1}$$  \hspace{1cm} (1)
Here $n_{\text{eff}}$ and $\frac{dn_{\text{eff}}}{d\lambda}$ denote the effective index of refraction and its first order dispersion for a cavity mode at the wavelength $\lambda$, respectively. The term $\left(n_{\text{eff}} - \lambda \frac{dn_{\text{eff}}}{d\lambda}\right)$ defines the group index of refraction, $n_g$, which was calculated for all confined optical eigenmodes by using a finite-difference-time-domain (FDTD) solver from the Lumerical Inc (see Supporting Information VI). The experimental modal pitch versus $1/L$ shown in Figure 4a can be well reproduced by the fundamental HE$_{11a/b}$ modes. As can be seen, most of the dominant PL undulations were resolved in the HE$_{11a}$, owing to its higher reflectivity and thus better modal quality factor than HE$_{11b}$ (see Supporting Information VII). The HE$_{11a}$ and HE$_{11b}$ modes are known to yield orthogonal emission polarizations that are parallel and perpendicular to the wire axis, respectively. Therefore, on the basis of the observed PL polarization, lasing from the NW1-3 is identified as originating from the HE$_{11a}$ mode.

A deep understanding on the efficiency of GaNAs as an optical gain material is crucial for realization of NIR NW lasers with superior device performance. For this purpose, we calculated the theoretical $g_{\text{th}}$ for all allowed cavity modes inside the nanowire, which can be expressed as,

$$g_{\text{th}} = \left(\frac{1}{R}\right) \left[\alpha_i + \left(\frac{1}{2\ell}\right)\ln\left(\frac{1}{R^2}\right)\right]$$

(2).

Here, $R$ is the reflectivity of the NW end facets, $\ell$ and $\alpha_i$ represent the mode confinement factor and internal propagation loss of the cavity mode, respectively. Wavelength dependences of these factors governing $g_{\text{th}}$ are provided in Supporting Information VII. Figure 3b shows a plot of the calculated $g_{\text{th}}$ over the spectral range of interest. As is clearly seen, the HE$_{11a}$ mode
possesses the lowest $g_{th}$ among all the other cavity modes above 970 nm, implying its favorable condition for lasing as optical pumping increases. Lasing in the HE_{11a} mode for the emission at 1000 nm can be understood by considering a simulated intensity profile for this centralized mode, as shown in Figure 3c-f for summed and individual polarization components. At this or longer wavelengths, the higher-order modes become more spread out of the NW cavity, which results in increased propagation loss. At the same time, their modal overlap with the GaNAs shell reduces. Both of these effects contribute to an increased threshold observed for the TE_{01} and HE_{21a/b} modes seen in Figure 3b. On the other hand, the mode - gain coupling is enhanced for the fundamental HE_{11a/b} modes, as the mode profile now extends toward the shell region, rendering the lowest threshold. It should be noted that the theoretical $g_{th}$ of HE_{11a} within the lasing range of 950 – 1000 nm has a value of around 3500 – 4000 cm⁻¹, which is lower than that extracted from the experimental power-dependent measurements. This discrepancy can be attributed to multiple factors including NW’s tapering and nonperfect end facet, which introduce additional scattering and diffraction loss. We simulated the material gain as a function of $P_{exc}$ for NW1 and the results are shown by the solid lines in the lower part of Figure 3b (see also Supporting Information V). Under increasing $P_{exc}$, the photoexcited carriers/excitons start to populate the energy band continuum, leading to a rising gain peak at 1.185 eV at 5 K. The gain spectrum reaches the threshold defined by HE_{11a} at 1022 nm, under a photo-excited carrier density, $n_c$, of ~ $4.0 \times 10^{18}$ cm⁻³. This wavelength is close to the observed lasing peak at 1000
nm. The good agreement between the experiment and simulation verifies the validity and accuracy of our analysis.

The lasing emission from GaNAs-based core/shell/cap NWs can be observed up to ~ 100 K with an increasing $P_{th}$ from 16 (5 K) to 29 (100 K) µJ cm$^{-2}$ per pulse, as shown in Figure 4. Further optically pumped lasing at higher temperatures is constraint by the output limit of our pulsed laser. The temperature-induced bandgap reduction, and hence the red-shift of the gain profile, causes the gradual transition of the dominant lasing intensity from the high-energy longitudinal modes to the lower ones. The measured pump threshold, $P_{th}$, as a function of temperature is plotted in the inset of Figure 4 and is fitted by an exponential function $P_{th} \propto e^{T/T_0}$, from which the characteristic temperature, $T_0$, of 160(±10) K is extracted. It is noteworthy that the derived $T_0$ is comparable with the reported values of 100 – 140 K for room-temperature lasing III-V NWs [7, 12] and is much higher than the value of $T_0 = 90$ K previously reported in the bare GaAs/GaNAs core/shell NWs without surface treatment [41]. The improved thermal stability of $P_{th}$ in the wires studied in this work is mainly attributed to surface passivation of the GaNAs shell by the GaAs cap layer, which effectively hinders non-radiative recombination (see also Supporting Information IX).

In conclusion, lasing at 1µm was demonstrated from the GaAs/GaNAs/GaAs core/shell/cap NWs containing only 2.5 % of N. The dominant lasing mode was identified as being the fundamental HE$_{11a}$ mode with the far-field polarization along the NW axis. Based on the experimental results complemented by the rate equation analysis, the threshold gain ranging
between 4100 – 4800 cm\(^{-1}\) was derived, with the highest spontaneous emission coupling factor of 0.8. The gain profile of the GaNAs active medium exhibits a large wavelength tunability achieved via alloying with only several percentage of N. The observed thermal robustness of lasing is enhanced by passivating the GaNAs shell with the GaAs cap layer. Our results, therefore, present an important progress towards the realization of telecom-wavelength operating NIR NW lasers based on dilute nitrides.

**Methods**

The GaAs/GaNAs/GaAs core/shell/cap NWs were grown by plasma-assisted Ga-catalyzed molecular beam epitaxy on (111) Si substrates. The NW growth involved several steps. Firstly, the GaAs core was grown via vapor-liquid-solid growth at 580 °C for 30 mins, with an equivalent pressure of an As\(_4\) beam of 1 × 10\(^{-3}\) Par. After that, the growth was interrupted for crystallization of the Ga catalyst on top of NWs. Then Ga flux was reduced to 0.5 ML to launch the lateral growth of the GaAs core for another 20 mins. Next, the growth temperature was reduced to 500 °C and the nitrogen plasma was ignited to grow the GaNAs shell for 30 mins. Finally, an outmost GaAs layer was deposited for 30 mins to cap the GaNAs shell. The so-obtained NWs consequently possess the GaAs/GaNAs/GaAs core/shell/cap structure, with the total diameter of 400 nm and length of ~ 5 \(\mu\)m. The crystalline structure of the NWs is dominated by the zinc-blende phase (~ 80%) but has inclusions of the wurtzite phase (~ 20%). Further details on the growth method and structural quality of the NWs can be found in Ref. 48.

\(\mu\)-PL measurements were performed in a backscattering geometry in a He-flow cryostat. For single wire measurements, the NWs were mechanically transferred to a gold substrate. A Ti: sapphire laser with an output wavelength at 780 nm was used as an excitation source. The
laser was operated in a continuous-wave mode to measure temperature-dependent PL spectra, and then switched to a pulsed mode, with a repetition rate of 76 MHz and a temporal pulse width of 150 fs, to perform power-dependent PL measurements. The excitation laser and the PL emission were guided through a 50×/NA = 0.5 microscope objective, with a focused spot size of 25 μm in diameter. PL signal from the NWs was dispersed by a single grating monochromator and detected by a charge-coupled-device camera. The PL polarization was resolved by means of a linear-polarization analyzer, which comprised of a rotatable half-wave-plate and a fixed Glan-Thompson linear polarizer.

ASSOCIATED CONTENT

Supporting Information.

FDTD simulations of the threshold gain for different optical modes as a function of emission wavelength, the rate equation analysis. This material is available free of charge via the Internet at http://pubs.acs.org

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REFERENCE


Figure 1. (a) The SEM image of a single transferred GaAs/GaNAs/GaAs core/shell/cap NW on a Au substrate, labeled as NW1. (b) Room-temperature spatially resolved $\mu$-PL spectra of NW1, which were recorded at the positions indexed from “1” to “11” in (a). All spectra are vertically shifted for clarity. The inset sketches the cross-sectional structure of the investigated GaAs/GaNAs/GaAs core/shell/cap NW. (c) Temperature-dependent PL spectra of NW1. (d) PL intensity mapping (normalized to peak intensity) as a function of temperature based on (c). The PL peak positions in spatially-resolved and temperature-dependent PL measurements are tracked by the dashed arrows.

Figure 2. (a)-(c) Power-dependent PL spectra of NW1-3 at 5 K. Their corresponding mapping of the normalized PL intensity versus pump fluence is displayed in (d)-(f). (g) Integrated PL intensity as a function of $P_{\text{exc}}$ (i.e. the light-in and light-out plots) of the main lasing lines ‘i’-‘iii’ from NW1-3. The filled symbols represent the experimental data whereas the solid lines are the best fit to the data based on the rate equation analysis. The FWHMs of the PL signals ‘i’-‘iii’ versus pump fluence are shown by the open symbols. The inset in (b) shows PL polarization in spontaneous (open circles) and lasing (dots) regimes. Mode spacing for the NW1-3 with different wire lengths is marked by the paired arrows.
Figure 3. (a) Mode spacing (symbols) versus inverse NW length, measured from individual NWs on an Au substrates, the stars represent the results taken from NW1-3. The solid and dashed lines are the calculated mode separation for HE_{11a} and HE_{11b}, respectively. (b) The values of the threshold gain calculated for the HE_{11a/b}, TE_{01}, and HE_{21a/b} modes over the spectral range of 850 – 1120 nm are shown by the symbols. The solid lines in the bottom part of the figure represent the material gain spectra in the GaNAs shell as a function of carrier density. (c-f) The HE_{11a} mode intensity and its polarization components along the x, y and z axis, respectively. The cross-sectional area of the GaNAs shell is outlined by the dotted lines.

Figure 4. Temperature-dependent PL spectra in the lasing regime. The inset shows the pump threshold, $P_{th}$, as a function of temperature, which was fitted by the exponential function $P_{th} \propto e^{T/T_0}$ (the dashed line) with the characteristic $T_0$ of 160(±10) K.
Figure 1
Figure 2
Figure 3
Figure 4